

REMARKS

Claims 1, 3-8 and 11 are all the claims pending in the application.

Page 2 of the Office Action states that the Examiner has withdrawn all the previous prior art rejections. However, the Examiner has maintained all the rejections, and has newly rejected claim 11, as discussed below.

The Examiner indicates that claim 11 was inadvertently identified in applicant's previous response as being "previously presented," rather than "new."

Since the Examiner has treated claim 11 on the merits in the present Office Action, applicant has indicated in the above Listing of Claims that claim 11 is identified as "previously presented", and thus no correction should be necessary.

Claims 1, 4, 5, 6, and 8 remain rejected under 35 U.S.C. § 103(a) as being unpatentable over Kitagawa et al (Microstructures and Thermoelectric Properties of $(\text{FeSb}_3)_{1-x}\text{La}_x$ Ribbons) in view of Lange (Lange's Handbook of Chemistry, Table 3.2) and Knosp et al (US 20020037453).

Applicant submits that Kitagawa et al, Lange and Knosp et al do not disclose or render obvious the subject matter of claims 1, 4, 5, 6 and 8 as amended above and, accordingly, request withdrawal of this rejection.

Applicant has amended claim 1 to recite that the alloy contains a $\text{RT}_4\text{Sb}_{12}$ composition, and that the alloy contains a filled skutterudite phase in an amount of at least 95 mass %. Support for these amendments can be found at page 6, lines 14 to 17 and page 7, lines 25 to 26.

The Examiner relies on Kitagawa et al for disclosing a filled skutterudite structure comprising a skutterudite-based alloy that is melted at a temperature sufficient to melt at least iron (Fe), and is subsequently spin-casted. The Examiner refers to page 334, second column, first paragraph of Kitagawa et al. The Examiner now agrees that the spin-cast method of

Kitagawa is not the strip-casting method of the present invention. Knosp et al is cited and is relied upon by the Examiner for a teaching the claimed cooling rate during strip casting or spin casting.

Claim 1 has been amended to indicate that amount of a filled skutterudite phase in an solidified alloy is at least 95 mass% in the state where a melt has been rapidly quenched through strip casting to form the solidified alloy, namely, in the as-cast state of the solidified alloy.

It is self-evident that the spin cast alloy of Kitagawa et al is amorphous. Applicant encloses a copy of pages 863 to 866 of "Encyclopedia of Metallic Material," 1999, together with a partial translation thereof. In lines 12 - 13 of the left column, p. 864 of the reference "Encyclopedia of Metallic Material" (page 2 of the translation), it is described that an amorphous thin ribbon measuring about 200 mm in width and about 30 μm in thickness is produced by the spin cast method. Since the ribbons solidified by Kitagawa et al have a thickness of 20 μm ("2. Experimental"), they are amorphous in the as-cast state. In "2. Experimental" of Kitagawa et al, it is described that the ribbons were crystallized by a heat treatment carried out at a temperature of 973K or 773K for a time of 36 ks, namely 10 hours.

In contrast, the present invention cools the melt within a range of the temperature of the melt and 800°C at a rate of 5×10^2 to 3×10^3 °C/second by using the strip cast method. Since this method enables the amount of the filled skutterudite phase in the solidified alloy kept in the as-cast state to reach a level of 95 mass% or more, the present invention possesses the merit of obviating the necessity of a heat treatment.

Further, since Kitagawa et al do not describe a strip casting process, the combination of the Kitagawa et al spin casting with the cooling rate of the Knosp et al spin casting would not result in the present invention.

Still further, there are differences between the alloy disclosed in Knosp et al as compared to that of Kitagawa et al. Knosp et al disclose an alloy with a crystal structure having the formula CaCu_5 , (see, for example, Knosp et al at paragraph [0012]), whereas Kitagawa et al disclose a filled skutterudite. As disclosed in the present specification, a skutterudite has a crystal structure of $\text{T}(\text{Pn})_3$, and a filled skutterudite has interstitial spaces filled with a rare earth metal R. Since the crystal structure of the alloys of Kitagawa et al and Knosp et al are different, one of ordinary skill in the art would not have been led to combining the teachings of the Kitagawa et al and Knosp et al.

The Examiner, in the Office Action, at page 4, lines 6 - 9, asserts that Knosp et al teach in paragraph [0009] an alloy comprising a rare earth metal, a transition metal, and metallic antimony, and asserts that this alloy is melted and rapidly cooled using either a melt spinning process or strip casting at a cooling rate of 10°C/s to 10^{60}C/s as disclosed in paragraphs [0034] and [0036] of Knosp et al.

The alloy disclosed by Knosp et al, however, is an alloy of CaCu_5 crystal structure, namely, a hydridable alloy, that is used in the cathode of a "nickel-metal hydride (Ni-MH) storage cell". See paragraphs [0003], [0005], [0012] and [0038] of Knosp et al. In short, it is not the filled skutterudite-type alloy of this application.

The alloy disclosed in paragraph [0009] of Knosp et al is a hydrogen-absorbing alloy that is claimed in U.S. Pat. 5,753,386 to Inaba et al, which is cited by Knosp et al in paragraph [0009], and is not the alloy of the Knosp et al invention.

The composition of the alloy that is claimed in Knosp et al is represented by the chemical formula, $\text{M}_m\text{Ni}_a\text{Mn}_b\text{Al}_c\text{Co}_d\text{Cr}_e$. See paragraph [0012] of Knosp et al. Since M_m is a mischmetal consisting of a mixture of La, Ce, Nd, as disclosed in paragraph [0024] of Knosp et al, the alloy

claimed in Knosp et al differs from the hydrogen-absorbing alloy claimed in Inaba et al in respect that it does not necessitate the group consisting of the elements of Y, Fe, Cu, B, Si, S, Ga, Ge, Mo, Ru, Rh, Pd, Ag, In, Sn, Sb, Bi, P, V, Nb, Ta, and W.

The hydrogen-absorbing alloy claimed in Inaba et al is preferably heat treated in a temperature range of from 500°C - 900°C for 2 - 15 hours, as disclosed at column 11, lines 7-17 of Inaba et al. In contrast, the alloy claimed in Knosp et al is heat treated (annealing) in a temperature range of from 900°C to 1100°C for less than or equal to 16 hours. See paragraph [0032] of Knosp et al.

Thus, the alloy claimed in Knosp et al differs from the alloy disclosed in paragraph [0009] of Knosp et al (i.e., the alloy claimed in Inaba et al) in terms of composition and conditions of fabrication. Consequently, the alloy disclosed in paragraph [0009] of Knosp et al (i.e., the alloy claimed in Inaba et al) would not necessarily be manufactured even under the conditions of manufacture described in paragraphs [0034] and [0036] of Knosp et al, and claimed in Knosp et al.

As discussed above, the Examiner, at page 4, lines 6 - 9 of the Office Action, asserts that Knosp teaches an alloy comprising a rare earth metal, a transition metal, and metallic antimony that is melted and rapidly cooled using either a melt spinning process or strip casting at a cooling rate of 10°C/second to 10⁶°C/second. This assertion is in error because the composition of the alloy claimed in Knosp et al is represented by the chemical formula, $M_mNi_nMn_pAl_cCo_dCr_e$, and does not include antimony (Sb).

Further, Knosp et al nowhere specifically disclose the range of 5×10^{20} °C/sec to 3×10^{30} °C/sec with any casting, and do not disclose this range in connection with *strip-casting*. Examples 1 to 3 of Knosp et al disclose a cooling rate of 10°C/sec for forming a plate in a copper

mold, which is well below the presently claimed range. Higher rates of cooling of 10,000 to 100,000°C/sec are disclosed in connection with *atomization* of the alloy in Examples 4 to 6, which are well above the presently claimed range.

Knosp et al disclose a cooling rate of the alloy over a very wide range of 10°C/s to 10⁶°C/s. Thus, Knosp et al only disclose the range of cooling rates that can be realized in a number of solidification methods, but do not disclose the cooling range of the present claims for strip-casting.

Knosp et al neither disclose the range of temperature of the alloy during rapid cooling of a strip-cast method, nor disclose rapidly cooling between the temperature of the melt and 800°C. Further, Knosp et al neither disclose nor suggest the advantages obtained at a specific cooling rate.

Hence, Knosp et al nowhere disclose or suggest the claimed rate of cooling of the present claims in connection with strip casting.

As compared with Kitagawa et al and Knosp et al, the invention of claim 1 of the present application comprises controlling the cooling rate to within a temperature range from the temperature of the molten alloy to 800°C, to 5×10^2 to 3×10^3 °C/second in order to attain a filled skutterudite-type alloy. By cooling under the claimed conditions, it is possible to obtain an alloy texture formed of a uniform filled skutterudite phase. See the present specification, page 9, lines 16-22. It is also possible to obtain an alloy exhibiting a maximum peak intensity (namely, abundance ratio), attributed to the filled skutterudite phase of 95% or higher, even when the alloy as removed from a production apparatus employed in the SC (strip cast) process has not undergone any further heat treatment. See the present specification, page 10, line 16 - page 11, line 13.

The Examiner continues to rely on Knosp et al for a teaching of the interchangeability of either strip casting or spin casting as a method of quenching alloys. According to the Examiner, one of ordinary skill in the art would have recognized that the methods are able to be used as equivalents with a reasonable expectation of success.

Applicant submits that spin casting and strip casting are not necessarily equivalent or interchangeable as a method of quenching, and can produce different results. As evidence to show that spin casting and strip casting are not necessarily equivalent or interchangeable as a means of quenching and nevertheless can produce different results, applicant refers the Examiner to the enclosed partial translation of "An Encyclopedia of Metallic Material."

In particular, applicant refers the Examiner to the single roll spinning method depicted in Fig. 2(a) and the double roll spinning method depicted in Fig. 2 (b) at page 863 of "An Encyclopedia of Metallic."

As can be seen from the enclosure, a single roll spinning method is depicted in Fig. 2(a) and a double roll spinning method is depicted in Fig. 2 (b) at page 863 of "An Encyclopedia of Metallic Material." In lines 5 - 7 and lines 12 - 13 of the left column, page 864 (page 2 of the translation, it is described that the amorphous thin ribbon for use in the magnetic head is manufactured by the spin cast method.

A melt drag method depicted in Fig. 8 at page 865 and is a strip method. In lines 17 - 19 of the left column, page 865 (page 5 of the translation), the method is described to afford advantages as in finely dividing a texture.

Accordingly, it is understood that the spin cast method produces an amorphous alloy and the strip cast method produces fine crystals.

The Examiner continues to rely on Knosp et al for a teaching of a cooling rate for either of strip casting or spin casting that overlaps the claimed range.

The Examiner maintains the position that that the cooling rates taught by Knosp et al overlap with the instant range and discloses such a range in conjunction with strip casting (citing paragraphs [0034] and [0036]).

Knosp et al disclose a process for rapidly cooling its alloy at a rate of between $10^{\circ}\text{C}/\text{sec}$. to $1 \times 10^{6^{\circ}\text{C}}/\text{sec}$. See for example, col. 2, lines 15-19; col. 3, lines 54-58; col. 4, lines 6-12 and lines 61-63; and etc. However, as discussed above, Knosp et al nowhere specifically disclose the range of $5 \times 10^{2^{\circ}\text{C}}/\text{sec}$ to $3 \times 10^{3^{\circ}\text{C}}/\text{sec}$ with any casting, much less strip casting. Further, none of the examples in Knosp et al refers to strip casting, much less within the claimed temperature range. Moreover, Examples 1 to 3 of Knosp et al disclose a cooling rate of $10^{\circ}\text{C}/\text{sec}$ for forming a plate in a copper mold, which is well below the presently claimed range. Much higher rates of cooling appear to be described in connection with *atomization* of the alloy in Examples 3 and 4, which is well above the presently claimed range.

Applicant thus again submits that Knosp et al nowhere specify the claimed rate of cooling in connection with strip casting with sufficient specificity to have rendered obvious applicant's claims.

The Examiner further states that applicant has argued that "Since the crystal structure of the alloys of Kitagawa and Knosp are different, one of ordinary skill in the art would not have been led to combining the teachings of Kitagawa and Knosp" (pp 6 of Arguments).

The Examiner disagrees with this argument.

The Examiner states that Knosp et al teach an alloy comprising a rare earth metal, transition metal and metallic antimony (paragraph [0009]) that is melted and cooled via melt

spinning or strip casting. The Examiner states that one reading either the Knosp et al and Kitagawa et al references as a whole would understand that both references are directed to solving the same problem — quickly and effectively quenching an alloy. The Examiner asserts that, therefore, one of ordinary skill in the art at the time of the invention would have looked to the teaching of Knosp et al for methods of quenching an alloy.

Applicant believes that the Examiner may not have understood the argument that applicant presented in the previous Amendment that Knosp et al nowhere disclose that that an Sb containing alloy can be treated either by melt spinning or strip casting. Thus, paragraph [0009] of Knosp et al, which refers to an Sb alloy, is not the alloy that is described in paragraphs [0034] and [0036] of Knosp et al as being the alloy which can be treated either by cooling in a metal mold, or by melt spinning or strip casting. The alloy in paragraph [0009] of Knosp et al is the alloy of Inaba et al.

Further, Knosp et al disclose a teaching of quenching a specific alloy, and nowhere disclose that this method can be applied to all alloys regardless of their composition.

FIG. 1 and FIG. 2 of the specification of the Inaba et al patent (U.S. Pat. No. 5,753,386) which is cited by Knosp in [0009] both depict the spin cast method.

In all the embodiments disclosed in Inaba et al, the melt was rapidly quenched and solidified by using either of the roll methods that are depicted in FIG. 1 and FIG. 2. Incidentally, in Comparative Examples 1-5 of Inaba et al, the obtained alloy specimens were heated at 1000°C for 10 hours to perform a homogenization heat treatment to the alloy specimens. See column 14, lines 36-38 and column 18, lines 46-49 of Inaba et al.

While Inaba et al disclose an alloy which contains Sb, the composition thereof was $\text{ANi}_{3.75}\text{Co}_{0.70}\text{Cu}_{0.17}\text{Mn}_{0.35}\text{Al}_{0.20}\text{Sb}_{0.03}$ as shown in Embodiment 18 and set forth in TABLE 1,

indicating that the Sb content was only $0.03/6.20 \times 100 = 0.48\%$. In contrast, the alloy composition of Kitagawa et al is $\text{LaFe}_4\text{Sb}_{12}$ ("1. Introduction") and the content of Sb is $12/17 \times 100 = 70.6\%$.

Further, the alloy of Inaba et al and the alloy of Kitagawa et al are different in respect that the former is a hydrogen-absorbing alloy (ABSTRACT) and the latter is a thermoelectric conversion material ("1. Introduction"). Still further, the alloy disclosed by Knosp et al is a hydrogen absorbing alloy which contains absolutely no Sb.

As can be seen from the above, the alloy disclosed in Kitagawa et and the alloy disclosed in Knosp et al are such alloys that are different in composition and totally different in function.

The application of the strip cast method of Knosp et al to the alloy of Kitagawa et al, therefore, is not obvious.

The Examiner states that applicant argues that "the alloy claimed in Knosp differs from the alloy disclosed in paragraph [0009] of Knosp...the alloy disclosed in paragraph [0009] of Knosp...would not necessarily be manufactured even under the conditions of manufacture described..." (pp 7 of Arguments).

The Examiner responds by stating that Knosp et al is relied upon as a general teaching of methods of quenching an alloy. The Examiner states that Knosp et al is not relied upon to teach the compositional elements/crystal structure of the claims. The Examiner states that Kitagawa et al teach the filled skutterudite composition needed for the instant claim.

Applicant disagrees that Knosp et al disclose a general teaching of methods of quenching an alloy. As discussed above, Knosp et al disclose a teaching of quenching a specific alloy, and nowhere disclose that this method can be applied to all alloys regardless of their composition.

As can be seen from the above discussion, the cited documents do not render obvious the subject matter of the present claims.

In view of the above, applicant requests withdrawal of this rejection.

Claim 3 remains rejected under 35 U.S.C. § 103(a) as being unpatentable over Kitagawa et al (*Microstructures and Thermoelectric Properties of (FeSb₃)_{1-x}La_x Ribbons*) in view of Lange (*Lange's Handbook of Chemistry, Table 3.2*) and Knosp et al (US 2002/0037453) as applied to claim 1 above, and further in view of Hehmann et al (WO/199604409).

This rejection is essentially identical to the previous rejection over these references.

Claim 3 depends from claim 1. Accordingly, applicant submits that claim 3 is patentable for at least the reasons submitted above in connection with claim 1.

Also, Hermann et al (WO/199604409) disclose a method of using an inert gas only at pp. 31, section 1.1.2 and in the paragraph titled "Melt spinning (MS) and planar flow casting (PFC)" at pp. 70 (particularly at the first to second line from bottom of pp. 70).

Neither of the foregoing paragraphs of Hermann et al specifically disclose the upper limit and the lower limit of the pressure of the inert gas which is effective for curbing the amount of Sb during vaporization.

The alloy disclosed in Hermann et al is an anti-corrosion forged light alloy, such as an aluminum and magnesium alloy, but is not the type of alloy claimed (filled skutterudite-type alloy), and its composition differs from that of the alloy disclosed in Knosp et al.

The Examiner generally has not responded to the arguments that applicant previously submitted. Applicant submits that the Examiner is employing hindsight by relying on general teachings.

In particular, the Examiner has not responded to the arguments that Hermann et al do not specifically disclose the upper limit and the lower limit of the pressure of the inert gas which is effective for curbing the amount of Sb during vaporization.

In view of the above, applicant requests withdrawal of this rejection.

Claim 7 remains rejected under 35 U.S.C. 103(a) as being unpatentable over Kitagawa et al (*Microstructures and Thermoelectric Properties of $(\text{FeSb}_3)_{1-x}\text{La}_x$ Ribbons*) in view of Lange (*Lange's Handbook of Chemistry, Table 3.2*) and Knosp et al (US 2002/0037453) as applied to claim 5 above, and further in view of Hirota et al (US 6,322,637).

This rejection is essentially identical to the previous rejection over these references.

The Examiner asserts that it would have been obvious to one of ordinary skill in the art at the time of the invention to keep the oxygen, nitrogen and carbon content of modified Kitagawa's alloy at 0.2 mass% or less, as taught by Hirota et al at column 2, line 6 to column 3, line 7, to achieve desired design characteristics.

Claim 7 also depends from claim 1. Accordingly, applicant submits that claim 7 is patentable for at least the reasons submitted above in connection with claim 1.

Further, Hirota et al do not relate to a filled skutterudite, but relate to a 4-phase alloy containing an α -iron phase, an R-rich phase, an $\text{R}_x\text{T}_4\text{B}_4$ phase and a $\text{R}_2\text{T}_{14}\text{B}$ phase. Accordingly, one of ordinary skill in the art would not have any reason to combine Hirota et al with Kitagawa et al.

Additionally, Hirota et al do not disclose at column 2, line 6 to column 3, line 7, that their alloys include less than 0.2 mass% or less of oxygen, nitrogen and carbon. Hirota et al do not disclose the oxygen, nitrogen and carbon of their alloys.

Indeed, Hirota et al do not even disclose the amount of impurities contained in an alloy comprising a rare earth element R in combination with a transition metal element and boron B.

Further, the Examiner generally has not responded to the arguments that applicant previously submitted. Applicant submits that the Examiner is employing hindsight by relying on general teachings of the prior art.

In view of the above, applicant requests withdrawal of this rejection.

Claim 11 is newly rejected under 35 U.S.C. 103(a) as being unpatentable over Kitagawa et al. (Microstructures and Thermoelectric Properties of $(\text{FeSb}_3)_{1-x}\text{La}_x$ Ribbons) in view of Lange (Lange's Handbook of Chemistry, Table 3.2) and Knosp et al. (US 20020037453) as applied to claim 1 above, and further in view of Masumoto et al. (US 4572750).

Applicant submits that these references do not disclose or render obvious the subject matter of claim 11.

Claim 11 depends from claim 1 and further recites that “the receiving box is cooled at a rate of 2 °C/second at a temperature within the range of from 700°C to 500°C.”

When the cooling rate in the receiving box is adjusted, such as in an Ar atmosphere at atmospheric pressure, to 2°C/sec in a temperature range of 700°C to 500°C, the product has a filled skutterudite $[(\text{Ce}_x, \text{La}_{1-x}) \text{Fe}_4\text{Sb}_{12}]$ phase content of 99% or more, as determined by powder X-ray diffractometry. See page 14, line 29 to page 15, line 10.

The Examiner states that Kitagawa et al, Lange and Knosp et al are silent as to the receiving box being cooled at a rate of 2°C/sec at a temperature within the range of 700°C to 500°C.

The Examiner argues that it is known in the alloy preparation art to utilize a twice cooling method wherein the alloy is first quickly cooled and then at an order-disorder

transformation point the alloy is then cooled at a slower rate to achieve a preferred degree of order as taught by the newly cited Masumoto et al patent at column 2, line 44 to column 3, line 12. The Examiner states that the alloy of Masumoto et al is cooled to room temperature and therefore will also be cooled within the range of 700°C to 500°C.

The Examiner argues that it would have been obvious to one of ordinary skill in the art at the time of the invention to utilize a twice cooling method wherein the alloy is cooled at a slower rate (100°C/sec to 1°C/hour, which reads on the instant 2°C/sec) as taught by Masumoto et al in the preparation of the alloy as taught by modified Kitagawa et al to achieve a preferred degree of order in the alloy.

Further, the Examiner states that the use of refrigeration as a cooling method is well known to one of ordinary skill in the art. The Examiner argues that choosing a desired cooling method (such as refrigeration, or cooling a receiving box) would have been within purview of one of ordinary skill in the art.

Applicant submits that this rejection is based on hindsight. Matsumoto et al do not relate to cooling conditions from a melt. In Matsumoto et al, an already formed magnetic alloy is reheated to a temperature below the melting point and then subjected to cooling. Applicant submits that the method of Matsumoto et al has nothing to do with the method of the present invention.

Further, Matsumoto et al do not relate to a filled skutterudite. In addition, with respect to the Examiner's statement that the use of refrigeration as a cooling method is well known to one of ordinary skill in the art, applicant submits that this reasoning is based on hindsight. By the Examiner's reasoning, one could never obtain a patent that involves cooling.

Further, Kitagawa et al disclose, at lines 7 - 10 of "2. Experimental" in the right column of page 334, a procedure which anneals a melt after it has been solidified. The annealing contemplated by Kitagawa et al is to make the solidified melt maintained at a fixed temperature (700°C (973K) or 500°C (773K)) for a fixed time (1 hour (36 ks)).

Then, Knosp et al disclose in paragraph [0009], while citing U.S. Pat. No. 5,753,386 to Inaba et al, a method for fabricating an alloy including rapidly quenching the molten metal at a rate of at least 100°C/s and subsequently subjecting the rapidly-quenched alloy to a homogenizing heat treatment at a temperature from 500°C - 900°C. In the case of this homogenizing heat treatment, however, when the heat treatment time is shorter than 1 hour, the effect for removing the internal distortion in the alloy will be lowered. See column 11, lines 11-13 of U.S. Pat. No. 5,753,386 to Inaba et al.

Masumoto et al, solely in view of the substantial effects on the resultant magnetic properties of the alloy, offer an explanation that the cooling rate from the temperature for the solution treatment to the temperature above the order-disorder transformation point is irrelevant whether the cooling is quick or slow. See column 2, line 66 to column 3, line 3. In short, Masumoto et al do not disclose the necessity for adopting two kinds of method for cooling the melt, namely the method of cooling an alloy "by rapidly quenching it and subsequently slowly cooling," for the sake of obtaining an alloy texture formed of a specific homogenous phase.

As can be understood from the above, the procedure of rapidly quenching a melt till solidification and subsequently subjecting the solidified melt to a heat treatment is generally carried out in the prior art. The adoption of the two kinds of methods for cooling a melt as a starting material for the purpose of obtaining an alloy texture formed of a specific homogenous phase is not disclosed in Kitagawa et al, Knosp et al and Masumoto et al.

The present invention as set forth in claim 11, therefore, was not obvious to persons skilled in the art at the time of the present invention.

In view of the above, applicant requests withdrawal of the rejection of claim 11.

In view of the above, reconsideration and allowance of this application are now believed to be in order, and such actions are hereby solicited. If any points remain in issue which the Examiner feels may be best resolved through a personal or telephone interview, the Examiner is kindly requested to contact the undersigned at the telephone number listed below.

The USPTO is directed and authorized to charge all required fees, except for the Issue Fee and the Publication Fee, to Deposit Account No. 19-4880. Please also credit any overpayments to said Deposit Account.

Respectfully submitted,

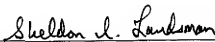
SUGHRUE MION, PLLC
Telephone: (202) 293-7060
Facsimile: (202) 293-7860

WASHINGTON OFFICE

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Sheldon I. Landsman
Registration No. 25,430